PREDICTIVE SURROGATE MODEL FOR NOx EMISSIONS IN GAS TURBINE SYSTEMS FED WITH AMMONIA/HYDROGEN BLENDS

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Abstract
Renewable energy sources must be widely integrated to face climate change. One important option that can be produced from surplus solar and wind energy is hydrogen, which may be used as an alternative fuel for technologies that are already in use. On the other hand, ammonia, that it is easily liquefied and distributed through the existing infrastructure, can act as an excellent hydrogen carrier.
This study aims to investigate the feasibility of using ammonia/hydrogen mixtures in gas turbines by applying numerical techniques with low computational cost. A chemical reactor network (CRN) based surrogate model is developed to estimate the effect of operational and model parameters on NOx emissions in a gas turbine fed with ammonia/hydrogen mixtures. To this purpose, we use Uncertainty Quantification (UQ) to calibrate the network and identify which operational and model qualities have the greatest impact on the estimation of NOx emissions.

Introduction
Gas turbines are an integral part of the global energy scenario, requiring initiatives to reduce carbon emissions and improve environmental sustainability. Hydrogen (H_2), derived from renewable energy sources, and ammonia (NH_3), used as its stable carrier, are among the leading alternative fuels under consideration due to their favorable storage characteristics. However, the different thermo-chemical properties of H_2 and NH_3 present unique challenges. Hydrogen is highly reactive compared to conventional fuels and has a high adiabatic flame temperature, which can result in increased thermal NOx emissions and flame instability (flashback). Conversely, ammonia is less reactive, characterized by low flame speeds that can lead to flame extinction (blow-off) and it can contribute to higher NOx production due to its fuel-bound nitrogen.
Considering these challenges, it is imperative to conduct comprehensive research on these fuels to assess their viability as substitutes for fossil fuels. Effective strategies like staged combustion and the blending of fuels, particularly blending ammonia with more reactive fuels such as methane or hydrogen, the latter obtained from ammonia cracking, have shown promise in improving the combustion characteristics of ammonia [1].
The investigation of new fuel mixtures through experimental setups or detailed numerical simulations using Computational Fluid Dynamics (CFD) can be

prohibitively expensive and time-consuming, given the need to investigate a wide range of conditions such as mixture composition, equivalence ratio, injection pressure, and temperature.
In this context, the Chemical Reactor Network (CRN) approach, first introduced by Bragg [2], emerges as a cost-effective and computationally efficient alternative. This methodology simplifies the representation of complex thermochemical and flow fields by dividing them into a network of discrete volumes modeled as canonical chemical reactors - typically Perfectly Stirred Reactors (PSRs) and/or Plug Flow Reactors (PFRs). These interconnected reactors exchange mass and heat and are designed to encapsulate the essential characteristics of the overall flow and thermos-chemical field.
However, the application of CRN to gas turbines is challenging, as it should replicate faithfully the intricate dynamics which is characterized by zones of varying temperature, flow, and chemical species concentration. Accurately modeling these complexities using zero-dimensional reactors that build the CRN is crucial.
Indeed, the effectiveness of the CRN model is highly dependent on the network configuration which should be devised by considering the flow features [3-4] as well as on model parameters as the chemical schemes. The latter may pose significant issues in case of novel fuels, because of the limited validation in literature.
The reliability of the approach is constrained by potential errors arising from network modeling and the calibration of model parameters. To address these limitations, Uncertainty Quantification (UQ) techniques are applied to quantify the propagation of uncertainties related to both operational and modeling parameters, and to assess the confidence level of the model. This type of analysis can be used to develop high fidelity and resolution models (surrogate models) based on the original CRN model, providing a useful tool for identifying the parameters that have a greater weight on the estimates of the quantities of interest analyzed and the relative range of interest (i.e., NOx and unburned NH_3).

Case study

The case study is the high-pressure generic swirl burner (HPGSB), housed within the high-pressure optical chamber (HPOC) [5]. The HPGSB uses quartz windows and a cylindrical quartz burner confinement to provide optical access to the flame. The HPGSB is modular and can be operated with a wide range of swirl numbers. Previous works [5-6] include further information on the design and operation of this high-pressure combustion rig test facility.

Numerical approach

The CRN code is written in Python using Cantera [7], an open-source software for thermodynamics and reaction kinetics. To investigate a fully CRN method without the use of CFD or other information, the CRN is developed using a simple one-stage configuration, with the flame zone as a PSR reactor and the post-combustion zone, characterized by unidirectional flow using a PFR reactor as shown in Figure 1.

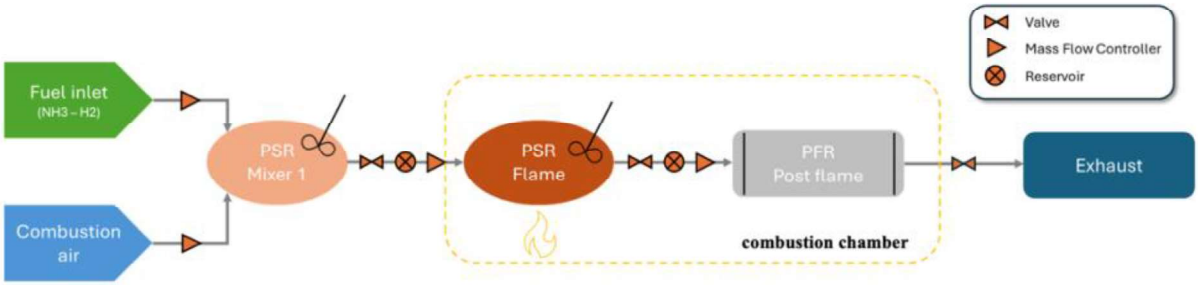

Figure 1. One stage Chemical Reactor Network structure.

Figure 1 exhibits an additional non-reactive PSR reactor (Mixer 1) that has the purpose of mimicking the mixing zone outside the combustion chamber for the characterization of premixed processes such as those being investigated on the HPGSB setup [4]. In this simple one-stage CRN, the model parameters are reduced to the kinetic mechanism used and the fraction of the flame volume V_F/V_T, which is the portion of the chamber occupied by the flame compared to the total of the chamber, with the mixer being an external and analytically defined element. In this scenario, we are investigated and quantified the uncertainty of operating and modeling parameters by assessing their impact on the selected quantities of interest, such as NOx and unburned NH_3.

The idea is to consider operating or modeling parameters not as deterministic variables, but rather through analytical sampling with an appropriate Probability Density Function (PDF), and to assess how uncertainty propagates at a computational level on the quantification of the desired outputs. This methodology can lead to direct calibration through comparison with experimental data or to a retro-calibration and CRN optimization phase, eliminating the parameters that do not influence the estimate of NOx emissions or unburned NH_3, optimizing the performance of the system and improving the confidence and robustness of the model. The propagation of uncertainty of the paraments is evaluated by developing a low-cost surrogate model that is trained on the outcomes of CRN simulations at specific points on a grid of size N, where N is the number of input parameters studied (N = 4).

Table 1: Operating and model parameters with their range.

Operating parameters	**Range**
Ammonia content NH_3 [%vol]	20 - 90
Equivalence ratio ϕ [-]	0.35 - 1.40
Inlet temperature T_{in} [K]	430 - 700
Model parameter	
Flame PSR volume fraction [$\%V_F/V_T$]	10 - 80

Table 1 presents the parameters and the respective distribution range. The surrogate model is developed through the advanced Monte Carlo approach [8], using a polynomial best-fit method, beginning from simulations of the original CRN at known quadrature points. These grid points are calculated using Newton Cotes quadrature formulas [8]. The surrogate model is a polynomial correlation with multiple variables (our operational and model parameters) that calculates our variable of interest, which in this case is NOx emissions in the parameter space, $NOx = f(\%NH3,\ \phi,\ T_{in}, \%V_F/V_T)$.
This model is used to conduct a global sensitivity analysis, determine the parameters that have the least influence on the CRN analysis, and generate response surfaces for NOx emissions.

Results: global sensitivity analysis
The global sensitivity analysis is conducted through the results obtained from our surrogate model in terms of PDFs and Sobol' indices of the quantity of interest, such as NOx emissions.

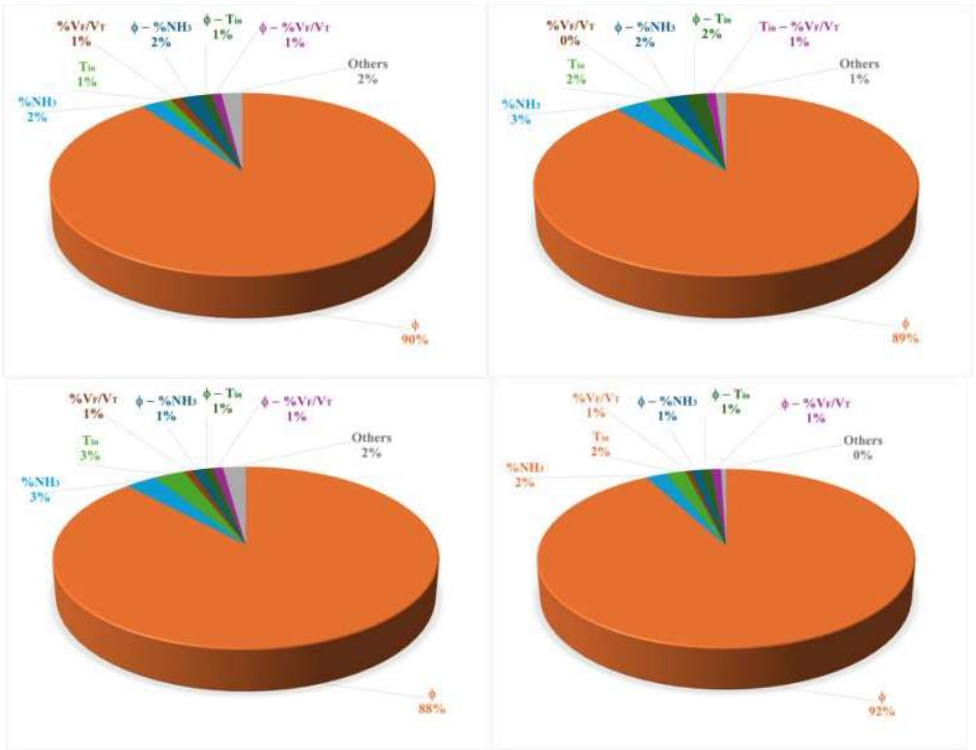

Figure 2. Sobol' indices for NOx emissions at different kinetic mechanism (a) Gotama et al. (b) Stagni et al. (c) Okafor et al. (d) Kaust et al.

These indices describe a variance-based statistical technique for global sensitivity analysis, which measures the individual importance of each parameter, as well as their joint effect on the quantity of interest, i.e., NOx emissions. In the first step, the impact of the kinetic mechanisms on the prediction of NOx emissions is evaluated

by analyzing four chemical schemes: Stagni et al [9] with 31 species and 203 reactions, Gotama et al [10] with 32 species and 165 reactions, Okafor et al. [11] with 59 species and 256 reactions and Kaust et al. [12] with 38 species and 263 reactions.

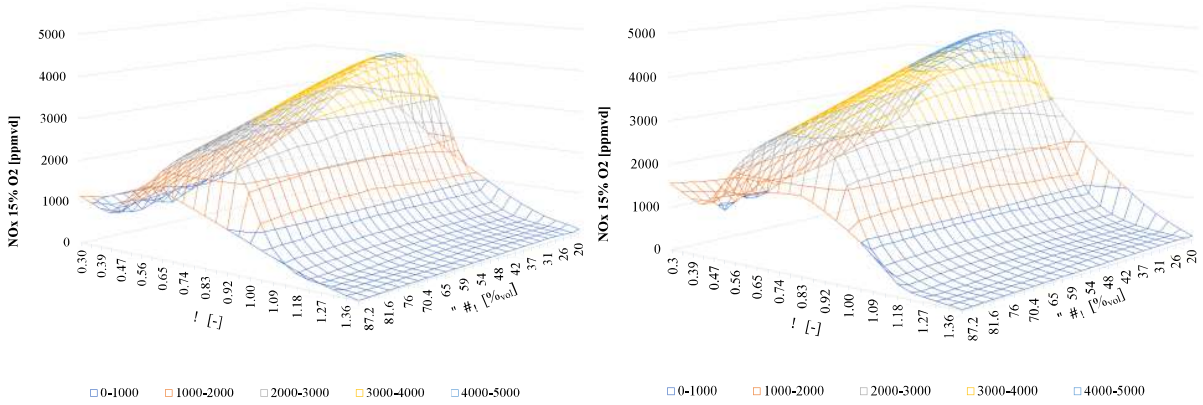

Figure 3. Maps (slice at T_{in} and $\%V_F/V_T$ constant) for NOx emissions at different kinetic mechanism (a) Gotama et al. [10] (b) Stagni et al. [9].

Figure 2 shows the Sobol' indices for NOx emissions by varying the kinetic schemes in the surrogate model. We can note that the equivalence ratio (ϕ) has the greatest influence on the estimation of NOx, with an index value equal to Iϕ = 90% -89%-88%-92% for the Gotama et al. [10], Stagni et al. [9], Okafor et al [11], and Kaust et al. [12] mechanisms respectively. The influence of the other parameters and their coupling is almost negligible. It should also be noted that the parameter related to the model (volume fraction of the flame reactor compared to the total volume of the combustion chamber) is 0%-1%, regardless of the kinetic mechanism analysed. This indicates that the influence of this parameter does not affect the estimation of NOx emissions compared to the other operating parameters. This does not mean that the estimation of the emissions is not quantitatively influenced by this parameter, but that by trying to characterize a wide range of configurations at the same time, the overall effect on the trend obtained is irrelevant. To visualize the trend of NOx emissions in a map, we consider a slice within the four-dimensional parameter space, fixing two parameters at a time, such as T_{in} and $\%V_F/V_T$, as shown in Figure 3, where only two kinetic mechanisms are considered (Stagni et al. [9] and Gotama et al. [10]). We can note that varying the kinetic schemes, the NOx emissions behavior agrees with each other, with a strong dependence on the equivalence ratio.

Conclusion

A surrogate model based on the single-stage CRN is developed to emulate the gas turbine system fed with ammonia/hydrogen mixtures. The surrogate model is used to perform a global sensitivity analysis of NOx emissions to operating and modelling parameters. The model allows the construction of operational maps of NOx emissions in parameter space. It also allows estimation of the effect of individual parameters and their coupling on the prediction of NOx emissions.

The surrogate model offers the possibility of providing concrete support for the definition of experimental setups, while highlighting the confidence level of the estimates and the robustness of such models.

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